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Explosive Characteristics of Cast PBX Based on HMX, Ammonium Perchlorate and Aluminium

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Explosive characteristics determination of cast explosives based on octogen (HMX), ammonium perchlorate, aluminium and polyurethane binder are presented. Shock sensitivity and PBX detonation parameters: detonation velocity, particle velocity, chemical reaction zone duration, chemical reaction zone width, polytrophic exponent of explosion products and detonation pressure are determined.

Key words: **cast explosives, explosive characteristics, explosive sensitivity, initiation, octogen, oxidizer, aluminium, ammonium perchlorate, detonation parameters.**

Abbreviations and symbols

HMX – octogen
PBX – polymer bonded explosives
Al – aluminium
AP – ammonium perchlorate
ZND model – Zeldovich-von Neumman-Döring model
CJ – Champan-Jouguet
d_m – average particle diameter
TDI – toluene diisocyanate
d charge diameter
D detonation velocity
d_f failure diameter
d_c critical diameter
Q – total chemical energy of decomposition
Q_D – detonation energy
P_c – critical shock pressure
l – attenuator thickness
TNT – trinitrotoluene
u_{CJ} – particle velocity
a – chemical reaction zone width
P_{CJ} – detonation pressure
ρ_e – explosive charge density
ρ_{CJ} – detonation products density
n – polytrophic exponent of explosive products
τ – chemical reaction zone duration

Introduction

POLIMER bonded explosives based on hexogene or octogene, aluminium and polymer binder (PBX) fall into castable secondary explosives [1-3]. Besides decreased detonation wave parameters, they are characterised by relatively slower delivery of energy in the detonation process and longer positive impulse phase. These explosives are used for the laboration of warheads with strong blast effect, such as main charge explosives for Navy underwater weapons (mines, torpedoes, bombs) and missile warheads.

Besides metal powder, the oxygen-enriched components (oxidizers) are added to these composites for the blast effect increasing. That will make the metal powder oxidation in the detonation wave more complete and the realisation of secondary, exothermic Al reactions in the Taylor wave. Ammonium perchlorate (AP) is significant as an ingredient due to its detonation and oxidizing properties. The most used is bimodal mixture of coarse (200 μm) and fine fraction (~10 μm) [4, 5].

As aluminised PBX are heterogeneous systems, their explosive decomposition is a very complex process which differs from classical detonation theory for condensed explosive materials. The detonation heat is separated in two steps – in primary decomposition of initial component and secondary, subsequent effect of Al with decomposition products, which increases total potential energy of the composite and, consequently, increases the blast effect.

The detonation mechanism of PBX based on ammonium perchlorate and aluminium can be described by ZND (Zeldovich-von Neumman-Döring) model for non-ideal explosives [5]. Two factors determine this non-ideal behaviour: relatively slow kinetics and an incomplete oxidizer decomposition (Al oxidation is reduced in stationary detonation zone) and secondary exothermic Al reaction with detonation products behind Chapman-Jouguet, *CJ*, point in Taylor rarefaction wave.

In principle, four steps which determine the detonation process can be separated:

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1. the octogen (HMX) decomposition according to the reaction:

$$C4H8O8N8 \rightarrow 4\ N2 + 4\ H2O + 2\ CO2 + 2C; \qquad (1)$$

2. the binder decomposition;
3. ammonium perchlorate decomposition according to the reaction (complete decomposition):

$$2\ NH_4ClO_4 \rightarrow 4\ H_2O + N_2 + Cl_2 + 2\ O_2, \qquad (2)$$

or according to reaction (incomplete decomposition – Al melting and ignition):

$$2\ NH_4ClO_4 \rightarrow 3\ H_2O + N_2 + 2\ HCl + 2{,}5O_2; \qquad (3)$$

4. the interaction among the above ingredients decomposition products and Al combustion in the detonation product flow, followed by oxidation reactions:

$$2\ Al + 3\ CO_2 \rightarrow Al_2O_3 + 3\ CO, \qquad (4)$$

$$2\ Al + 3\ H_2O \rightarrow Al_2O_3 + 3\ H_2. \qquad (5)$$

The illustration of chemical decomposition process for this type of non-ideal explosive is presented in Fig.1.

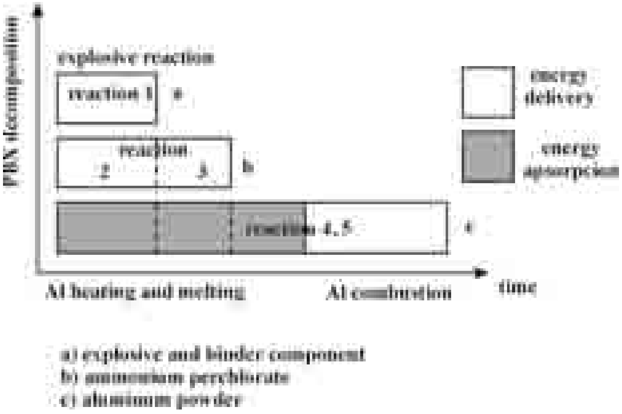


Figure 1. Schematic review of PBX decomposition

Based on results from [5, 6], the decomposition of HMX and polymer binder occurs completely in the steady-state zone. The behaviour of the oxidizer and Al depends on the particle dispersion and the mechanism of metal powder combustion in the PBX detonation products flow. It was established that fine grain-size ammonium perchlorate (10 µm) completely decomposes in the steady-state detonation zone. In the case of coarse grain-size AP (200 µm) the decomposition can be prolonged, for a few microseconds in the non steady-state zone. The PBX detonation parameters depend on whether or not the thermodynamic equilibrium of Al oxidation forms in the chemical reaction zone and gasodynamic equilibrium of decomposition products is attained. The equilibrium can be accomplished only if there is an adequate size of explosive particles filler.

The results of research of PBX based on HMX, with 20 - 35 mas. % of ammonium perchlorate and the examination of the influence of the mass ratio on PBX explosive characteristics are presented in this paper.

Experimental results and discussion

PBX processing

The PBX processing is realised by using an optimized composition of a non-energetic polymer binder, bimodal mixture of HMX crystals, modern equipment (Planetron HKV vertical mixer) and full control of technological parameters and the remote control of processing [7]. The following components are used:

bimodal mixture of coarse (200 µm) and fine (10 µm) ammonium perchlorate in mass ratio 60:40 (AP-I) and 20:80 (AP-II),

bimodal mixture of coarse (200-500 µm) and fine (50-150 µm) octogene, mixed in mass ratio 70:30,

aluminium powder with mean diameter of 15µm,

polyurethane binder, synthesized by polycondensation of toluene diisocyanate (TDI-80) and liquid polyether poliol [8].

A few PBX explosive compositions (PBX-1 to PBX-4) with 20 % of polyurethane binder, 25 % of Al and 25-35 % of AP were cast by using the HMX with determined granulations. Compositions were characterised by measuring of detonation wave parameters and by determining the shock sensitivity.

PBX detonation velocity and detonation critical diameter

The detonation velocity was measured in the steady-state detonation zone, in charges of 30, 40 and 50 mm in diameter, by using ionisation gages and an electric counter (method SNO 1475). Charges were boosted by FH-5 (ρ_0 = 1.60 g/cm^3).

The critical diameter of PBX was also determined.

The PBX composition and measured explosive characteristics in the function of particle size distribution and the AP content are presented in Table 1.

The detonation parameters' experimental values are the average values based on 3 experiments. The relative errors of measurements are between 1 and 2 %.

The functional dependences $D = f(mas.\%\ \text{AP})$ and $D = f(d)$ are presented in Figures 2-3, respectively. The influence of oxidizer dispersion on PBX detonation characteristics was examined by using the composition PBX-3. The mark * in Table 1 designates the composite with granulation AP-II; all other compositions were cast with AP-I.

Table 1. PBX composition and characteristics

Explosive	composition (mas.%) (HMX/AP/Al/PU)	*d* (mm)	D_{mean} (m/s)
PBX-1	(55/-/25/20) ρ = 1.668 g/cm³	30	7396
		40	7336
		50	7342
PBX-2	(35/20/25/20) ρ = 1.714 g/cm³	30	6067
		40	6217
		50	6434
PBX-3 PBX-3*	(25/30/25/20) ρ = 1.717 g/cm³	30	5725
		40	5888
		40*	5894
		50	5937
		50*	6003
PBX-4	(20/35/25/20) ρ = 1.729 g/cm³	30	charge was not initiated
		40 (d_f)	failure of detonation
		40	1758
		40	2353
		50 (d_c)	5450

It is evident from Table 1 that there is a reduction of detonation velocity in spite of charge density increasing. The detonation velocity linearly decreases with increasing of AP mass part. This is the result of the decreased part of explosive component, crystal HMX, on account of inert AP and Al adding in PBX (Fig.2).

The complete detonation was realised with the PBX-1 composition; in spite of charge diameter change the detonation velocity had the same value (Fig.3). The results are in good agreement with the results of aluminised PBX examination presented in [9].

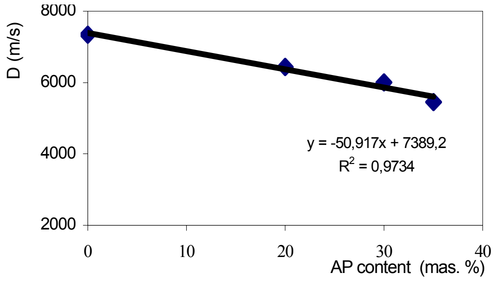


Figure 2. $D = f$(mas.% AP) dependence

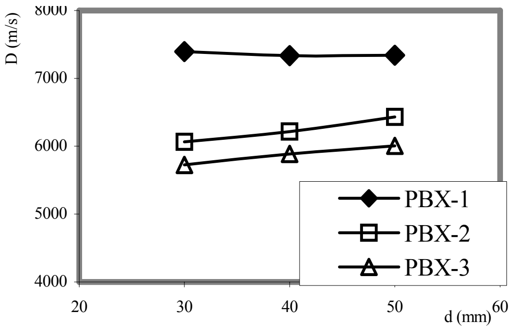


Figure 3. $D = f$(d) dependence

There is an increase in the detonation velocity of PBX-2 (with 20 mas.% AP) with charge diameter increasing. Detonation velocity increases ~ 200 m/s with increasing of the charge diameter of 10 mm. For this compound, the ideal detonation was not reached. The failure diameter is higher than 50 mm.

The detonation velocity increase is also evident for PBX-3, but the gradient increment is smaller: it is ~ 100 m/s. Like for PBX-2, the ideal detonation was not reached, and therefore the failure diameter is higher than 50 mm. AP dispersion change in the assigned variety interval of fine fraction did not affect the value of the detonation velocity.

Figure 4. Detonation failure in PBX-4

The composition of PBX shows typical non-ideal explosive behaviour. The test results show that the parameters of the established detonation strongly depend on the relation between HMX/AP and the charge radius. At *d*=30mm the initiation of the tested charge did not occur, and with PBX-radius charge *d*=40mm a non-stationary detonation process was initiated - in the first two tests very low detonation velocity was registered, and detonation interruption in the third test, Fig.4. Consequently, with the relation of ingredients HMX/AP =20/35 and A1 content of 25mas.%, mass part of non-explosive components is critical and the overall exothermal chemical reactions do not develop. Switching HMX with 35% AP has an inhibiting effect on the process of PBX decomposition and spreading of the detonation wave; therefore the complete conversion of PBX charge into detonation products is not evident.

These results are an experimental confirmation of the theoretical statements that the decomposition mechanism of the initial composition shows an important influence on the critical dimensions of the explosive charge, as well as conditions of induction and spreading of detonation. Depending on the kinetic characteristics of PBX ingredients, their mass ratio and other factors, different chemical and physical processes in detonation wave front can develop. For the realisation of composite explosive conversion all over the explosive charge to take place, due to the starting chemical reaction, the process has to be self-supporting, i.e. it has to have the proper kinetic characteristics. Since the tested PBX composites are non-homogenic, from the kinetic aspect, dismissing the fact that the metal powder and oxidizer supplement increase the total energy of the composite explosive, the larger part of that brisance energy [10] does not get released in a short period of time, which is characteristic for a chemical reaction zone. It is obvious that in PBX-4 compound with d = 40 mm the octogen content is not sufficient for the detonation wave to spread, there is a failure of the detonation process. These conclusions are supported by reference for compounds based on hexogen [11] presented in Table 2 and Fig.5. As the RDX content decreases in this series of materials from 100 % to 20 % by mass, the total energy increases, but the detonation energy decreases. In conditions of complete detonation, for compounds with 20 % RDX and 80 % non-ideal "inert" components, only about 17 % of the total energy contributes to the detonation process.

Table 2. Comparison of total and detonation energies [11]

Explosive	Total energy *Q* (KJ/kg)	Detonation energy *QD* (KJ/kg)	*Q/QD* (%)	*~dc* (mm)
RDX	5041.1	4777.4	94.7	2
PBXW-108 (RDX/HTPB=85/15)	5183.5	3697.1	71.3	10
PBXW-109 (RDX/Al/HTPB=65/20/15)	7892.5	3332.8	42.6	13
PBXN-111 (RDX/Al/AP/PU=20/25/43/12)	12246.9	1469.6	17.3	40

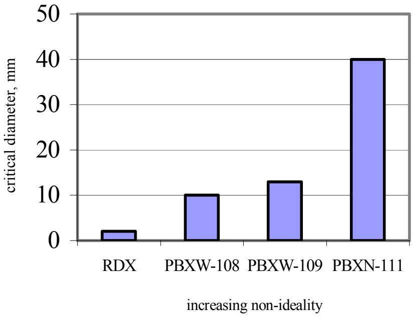


Figure 5. Critical diameter of detonation for different explosives [11]

For PBX-4 with charge diameter of 50 mm the steady-state detonation with the detonation velocity of D = 5450 m/s was registered. It means that for this compound failure and critical diameters are 40 mm and 50 mm, respectively (these values are higher than for PBX-1). It can be concluded from Table 2 and Fig.5 that the metal powder presence in PBX compounds increases the critical diameter, which is particularly shown in AP compositions.

PBX shock sensitivity

The GAP Test was used to determine the critical shock pressure [12]. The PBX-shock sensitivity determination was carried out by using the following system Φ = 40 mm:
the booster, FH-5,
the polyamide attenuator,
the tested PBX explosive charge ($\Phi = H$)
copper cylinder.

Critical initiation conditions were determined by a copper cylinder deformation.

The critical shock pressure value was calculated from the following expression:

$$P_{cr} = 203.32 \times e^{-72.284 \times l} \qquad (6)$$

where:
P_{cr} - the critical shock pressure (MPa),
l - the polyamide thickness (mm).

Results of the shock sensitivity determination for three explosive compositions: PBX-1, PBX-2 and PBX-3, are presented in Table 3 and compared with the conventional melt-cast TNT-based explosives (hexolite, octolite) [12,13].

Table 3. PBX and TNT-based explosive charges shock sensitivity results

Explosive	Granulation AP	Initiation	l (mm)	P_c (GPa)
PBX-1(without oxidizer)	-	+	34.8	1.64
PBX-2 (with 20 mas.% AP)	AP-I	+	33.1	1.86
PBX-3 (with 30 mas.% AP)	AP-I	+	28.0	2.67
PBX-3*(with 30 mas.% AP)	AP-II	+	28.9	2.52
Hexolite 60/40	-	-	-	1.95
Octolite 80/20	-	-	-	2.00
Note: * the same PBX composition, but different granulation of oxidizer (AP-II)				

Critical pressure values depend on the HMX/AP relation. Mass part of crystal octogen has dominant influence on the explosive behaviour under shock loading. Shock sensitivity is reduced with HMX content decreasing, i.e. increasing of ammonium perchlorate fraction.

Shock sensitivity does not decrease with the reduction of oxidizer particle size - both explosive compounds, PBX-3 and PBX-3*, have almost the same critical pressure values, although AP granulation differs.

Shock sensitivity of PBX with or without the oxidiser is at the shock sensitivity level of hexolite 60/40 and octolite 80/20.

PBX particle velocity

Particle velocity was measured using the electromagnetic method with the pulse magnetic field [14]. Results are oscilograms $u=f(t)$. Through analysis, the chemical reaction zone duration (τ) and the particle velocity at CJ state, u_{CJ}, were determined.

Results of particle velocity determination are presented in Table 4 and oscilograms for the tested compounds (PBX-1, PBX-2 i PBX-3) in Fig. 6. The detonation parameters experimental values are the average values based on three experiments. The relative error of the measurement is 2%.

Based on experimentally determined values for D, τ and u_{CJ} and hydrodynamic theory relations, other detonation parameters for Champan-Jougeut condition were calculated (Table 4):

- particle velocity, $u_{CJ} = 1.1 \dfrac{e_{CJ}}{B \cdot l}$ (m/s)

(coefficient 1.1 compensates the cable reducing), (7)

chemical reaction zone width,

$$a = \left(D - \frac{u_{CJ} + u_{\max}}{2} \right) \cdot \tau \text{ (mm)}, \qquad (8)$$

detonation pressure,

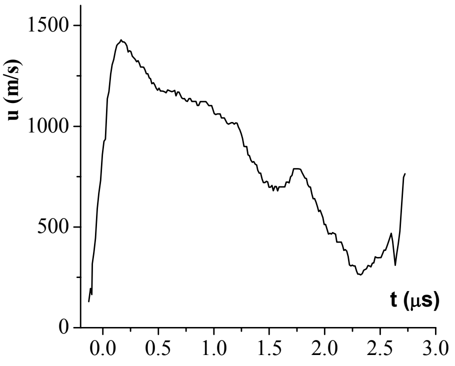
$$P_{CJ} = \rho_0 \cdot D \cdot u_{CJ} \text{ (GPa)}, \qquad (9)$$

detonation product density,

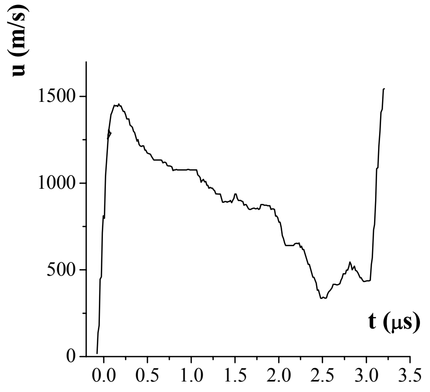
$$\rho_{CJ} = \rho_0 \cdot \frac{D}{D - u_{CJ}} \text{ (g/cm}^3\text{)}, \qquad (10)$$

polytropic exponent of explosive products,

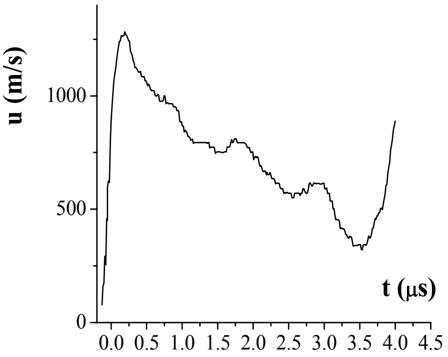
$$n = \frac{D}{u_{CJ}} - 1. \qquad (11)$$



a) PBX-1



b) PBX-2



c) PBX-3

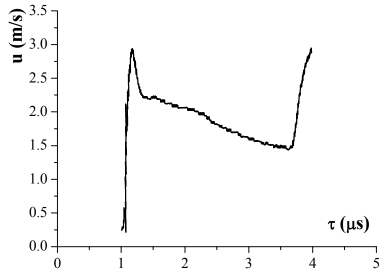
Figure 6. Electromagnetic gage records u = f(t) for the tested explosive compounds

Table 4. Results of PBX detonation parameter determination using the electromagnetic method

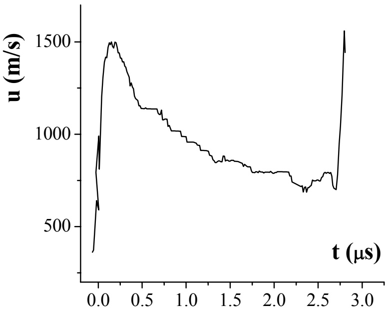
PBX (HMX/AP)	ρ_e g/cm³	D m/s	u_{CJ} m/s	τ μs	a mm	n	P_{CJ} GPa	ρ_{CJ} g/cm³
PBX-1 (55/**0**)	1.69	7358	1150.0	0.853	5.16	5.41	14.10	1.97
PBX-2 (35/**20**)	1.71	6434	1046.4	1.220	6.33	5.14	11.54	2.05
PBX-3 (25/**30**)	1.72	5937	850.3	1.640	7.88	6.01	8.67	2.00

By analyzing results of particle velocity measurement of composites it was concluded that the registered *u*(*t*) records are specific. The heterogeneity of the PBX composition is reflected in the structure of the detonation wave. Because of the metal powder and the oxidizer presence *u*(*t*) profiles for Al-PBX and Al-AP-PBX compounds have a characteristic shape which differs from *u*(*t*) for compounds without these ingredients. HMX-PU composite has triangular *u*(*t*) profile which is typical for high explosives [15].

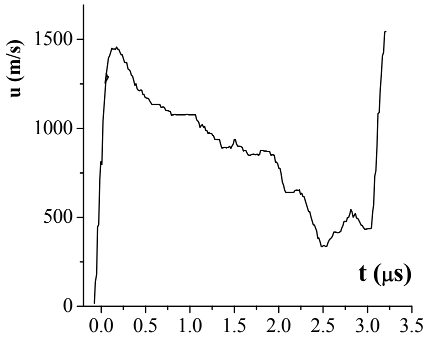
For clearer illustration of PBX compound influence, specific *u*(*t*) profiles for three different composites are presented in Fig.7. It can be noted that *u*(*t*) profile for PBX with Al addition (b) and Al-AP addition (c) does not have a sharp but somehow round von Neumman peak, (himpik) at the beginning of the *u*(*t*) record.



a) HMX-PU = 80-20



b) PBX-1



c) PBX-2

Figure 7. PBX compound influence on u (t) profile

However, sudden particle velocity decreasing behind the peak is evident, followed by slower decreasing with the plateau which is not characteristic for HMX compounds without aluminium (a).

Very specific *u*(*t*) records for composites with ammonium perchlorate were registered. All registered *u*(*t*) profiles had secondary plateau at the end of the chemical reaction zone. Three-level *u*(*t*) profiles can be attributed to the unbalance of PBX ingredients decomposition rates and change in kinetics of aluminium chemical reactions with detonation products. Registered *u*(*t*) records for PBX with 20 mas.% (PBX-2) and 30 mas.% (PBX-3) have no clear *CJ* point as in case of PBX-1. This suggests that all potential energy of the metal combustion is not realised in the detonation wave and that a part of Al reacts later on, behind the *CJ* point, in the non steady-state zone [12].

It can be concluded, based on the results from Table 4, that the particle velocity and the detonation pressure decrease with AP mass part increasing. Also, the oxidizer presence in these heterogeneous compounds increases the chemical reaction zone duration and width, which questions the hypothesis of some authors [10] that Al in chemical reaction zone reacts inertly.

The polytrophic exponent of detonation products, which has a value ~3 for conventional high explosives, is higher than for PBX without Al and it increases with the oxidizer content increasing.

Conclusion

From the research and results presented here, the following conclusions were drawn:

1. Composite explosives with different HMX/AP ratio, 25mas.% aluminium and 20mas.% polymer component were obtained by varying ammonium perchlorate mass percent from 20 to 35.
2. Compared to aluminised octogen compounds, compounds with ammonium perchlorate have lower shock sensitivity. Critical pressure values of the shock wave that initiate the examined composites are near the values for hexolite 60/40 and octolite 80/20.
3. The influence of the oxidizer particle size reduction on PBX behaviour under loading has not been demonstrated.
4. The detonation velocity linearly decreases with ammonium perchlorate content increasing although PBX density increases. AP granulation influence on the detonation velocity has not been shown.
5. The explosive component content is not the main indication of PBX detonability. Initiation and spreading of detonation in condensed explosives with oxidizers

strongly depends on the charge diameter. Increase in the detonation non-ideality and detonation velocity with the oxidizer content increasing is evident. The examined PBX with ammonium perchlorate show a very specific feature - the failure and critical diameter are higher and the detonation velocity is lower than for HMX-PU and HMX-Al-PU explosives.

6. The steady-state detonation was established only for PBX without ammonium perchlorate. The detonation velocity increases with charge diameter increasing for composites with 20mas.% and 30mas.% ammonium perchlorate. However, for these compounds an ideal detonation velocity was not reached because the failure diameter is higher than 50 mm. AP dispersion change in the assigned variety interval has no influence on the detonation velocity.
7. The compound with 35mas.% AP shows typically non-ideal behaviour. Detonation parameters strongly depend on the HMX/AP ratio and charge diameter. The failure and critical diameter for this compound are 40 mm and 50 mm, respectively.
8. The particle velocity u_{CJ} and the detonation pressure P_{CJ} decreasing with adding of aluminium and ammonium perchlorate to PBX was registered, along with decreasing of the duration and the width of the chemical reaction zone. Very specific $u(t)$ records registered for compounds with the oxidizer are the experimental evidence that metal powder in the presence of AP does not react like an inert material, but reacts with detonation products and partially oxidizes in the detonation wave front.
9. The presence of ammonium perchlorate in the composite explosive qualitatively changes the detonation wave front structure. The secondary plateau at the end of the chemical reaction zone was registered in PBX with an oxidizer using an electromagnetic gage (a compound without AP has only one plateau at the end of *CJ* plane). Three-level u (t) profiles can be attributed to unbalance of PBX ingredients decomposition rates and developing of Al exothermic reactions with decomposition products in the detonation wave front.
10. It can be concluded that the registered $u(t)$ profiles deviate from classical triangular profiles described by ZND model for high explosives but have characteristic, trapezoidal shape without clearly expressed von Neumman's peak. The changed profile structure and monotonous increasing of $u(t)$ behind *CJ* plane point to a detonation with more complex gasodynamics behind the detonation wave front and confirm additional energy release in the Taylor wave.

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Eksplozivne karakteristike livenih PBX na bazi HMX, amonijumperhlorata i aluminijuma

Prikazani su rezultati određivanja eksplozivnih karakteristika livenih eksploziva na bazi oktogena (HMX), amonijumperhlorata, aluminijuma i poliuretanskog veziva. Ispitana je osetljivost na inicijaciju udarnim talasom i određeni su detonacioni parametri PBX: brzina detonacionog talasa, brzina produkata detonacije, širina zone i vreme hemijske reakcije, stepen politrope, kao i pritisak detonacije.

Ključne reči: **liveni eksploziv, oktogen, oksidans, aluminijum, osetljivost na inicijaciju, parametri detonacije.**

Взрывчатые характеристики литых ПБЬ на базисе ХМЬ, аммония перхлората и алюминия

В настоящей работе показаны результаты определения взрывчатых характеристик литых взрывчатых веществ на базисе октогена (ХМЬ) аммония перхлората, алюминия и полиуретанового вяжущего вещества. Здесь тоже испытана и чувствительность на инициирование (возбуждение) детонационной волной и определены детонационные параметры ПБЬ: скорость детонационной волны, скорость продуктов детонации, широта зоны и время химической реакции, степень политропы, а в том роде и давление детонации.

Ключевые слова: литые взрывчатые вещества, октоген, характеристики взрывчатых веществ, чувствительность взрывчатых веществ, инициирование (возбуждение), окислитель, алюминий, аммония перхлорат, детонационные параметры.

Caractéristiques explosives des PBX de fonte à la base des HMX ammonium pechlorate et alumiium

Dans ce travail on a présenté les résultats de la détermination des caractéristiques explosives des explosifs de fonte à la base d'octogène (HMX), ammonium perchlorate, aluminium et liant polyuréthane. On a examiné la sensibilité quant à l'initiation de l'onde de choc. On a déterminé aussi les paramètres de détonation chez les PBX: vitesse de l'onde de détonation, vitesse des produits de détonation, largeur de la zone et le temps de la réaction chimique, degré de polytropie ainsi que la pression de la détonation.

Mots clés: **explosifs, octogène, propriétés des explosifs, oxydateur, aluminium,sensibilité des explosifs, initiation, ammonium perchlorate, paramètres de détonation.**